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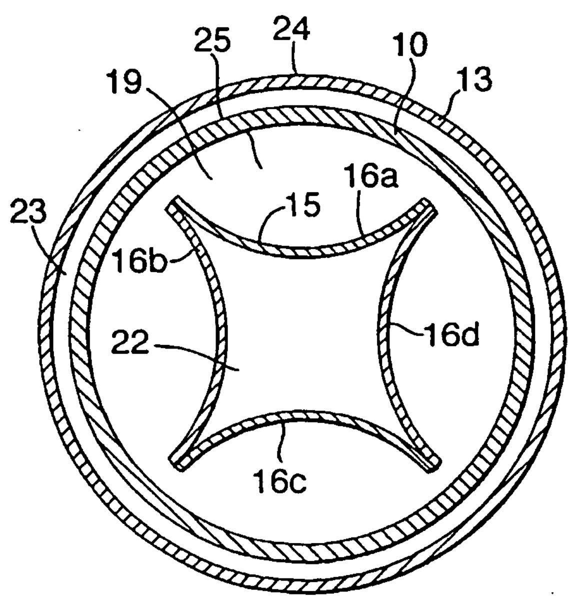
(54) Title: STEAM REFORMING APPARATUS



(57) Abstract: A reformer tube, for a steam reformer, having a double-tube configuration with an outer tube (10) of substantially circular cross section and an inner tube (15) of non-circular cross section disposed within the outer tube (10).

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Steam reforming apparatus

This invention relates to steam reforming apparatus and in particular to the design of steam reforming tubes.

In the steam reforming process a process gas, i.e. a mixture of a hydrocarbon feedstock and steam, and in some cases also carbon dioxide, is passed at an elevated pressure through catalyst-filled tubes which are externally heated by means of a suitable heating medium, generally a hot gas mixture. The catalyst is normally in the form of shaped units, e.g. cylinders, rings, saddles, and cylinders having a plurality of through holes, and are typically formed from a refractory support material e.g. alumina, calcium aluminate cement, magnesia or zirconia impregnated with a suitable catalytically active material which is often nickel or ruthenium.

In one form of reformer design, the reformer tubes are of a double-tube design, having an outer tube and a concentric inner tube thus providing a space between the inner and outer tubes. The catalyst units are disposed in this space, normally random packed as a result of pouring the catalyst units into the space from one end. In this arrangement, the outer tube is often closed at one end and the process gas passes through the space between the interior surface of the outer tube and the exterior surface of the inner tube, and then returns through the inner tube. An example of such a construction is described in US 4 690 690. The tubes normally have a circular cross-section and have a length of several metres, e.g. 5 to 15 m, and the outer tube typically has an outer diameter in the range 7 to 20 cm. The tubes are generally disposed vertically. In operation the tubes are heated to a high temperature, typically to a maximum temperature in the range 700°C to 900°C by a suitable medium flowing past the exterior of the outer tubes. This heating necessarily means that the tubes are subject to considerable thermal expansion, both longitudinally and radially, as the tubes are heated from ambient temperature at which the catalyst is normally loaded and the operating temperature and likewise to thermal contraction as the tube is cooled upon shut down of the process. The thermal expansion permits the catalyst units to settle down the space between the tubes and subsequent contraction, particularly that in the radial direction, subjects the catalyst units to severe crushing forces, often leading to catalyst breakage.

The circular cross-section of the outer tubes is dictated by the need for the tubes to withstand a substantial pressure differential between the pressure of the process gas within the tubes and the pressure of the heating medium. For example in many applications, the heating medium is at a pressure in the range 1 to 4 bar abs. while the process gas may be at a pressure in the range 20 to 80 bar abs. While this pressure differential may be decreased if the pressure of the heating medium is increased, for example where the heating medium is the process gas after further treatment, e.g. secondary reforming as described in the aforesaid US 4 690 690, there is still liable to be a pressure differential of several bar between the process gas and the heating medium.

On the other hand, where the process gas passes through the catalyst and then back through the inner tube, the pressure differential across the walls of the inner tube can be

relatively small and so the inner tube can be made from relatively light gauge material. We have now realised that because the inner tube does not have to withstand such large mechanical stresses as the outer tube, it need not be of circular cross-section and indeed advantages may result from the use of a non-circular cross-section.

Accordingly the present invention provides a reformer tube of the double-tube configuration having an outer tube of substantially circular cross-section and an inner tube disposed within the outer tube, said inner tube having a non-circular cross-section.

The invention also provides the use in a double-tube reformer of an inner tube having a non-circular cross-section.

As a result of the use of an inner tube of non-circular cross-section, the inner tube is more capable, than a tube of circular cross-section, of local deflection and so can deflect to accommodate settled catalyst units, thereby decreasing the crushing forces to which the catalyst units are subject.

One consequence of the use of an inner tube of non-circular cross-section within an outer tube of circular cross-section is that the width of the space between the tubes varies round the perimeter of the reformer tube, leading to differing radial depths of catalyst in this space. However, calculation has shown that such a variation in the depth of the catalyst space has negligible effect on the degree of reforming that will occur.

The cross-section of the inner tubes is non-circular: in particular the maximum cross-sectional dimension of the inner tubes is at least 1.2, and preferably at least 1.4, times the minimum dimension. The cross-section is preferably polygonal, particularly a regular polygon having 3 to 6 sides, preferably 4 to 5 sides. While the sides of the polygon may be straight, so that the cross-sections are those of e.g. an equilateral triangle, square, regular pentagon or hexagon, the sides of the polygon are preferably inwardly curved. The curve may be an arc of a circle or in particular is in the form of a catenary (hyperbolic cosine curve). In a preferred arrangement, the inner tube is formed by welding, e.g. by electrical resistance or laser seam welding, together the edges of metal strips of substantially uniform thickness that are curved across their width. The strips are preferably relatively thin; thus the maximum thickness of the walls of the inner tube, i.e. at the seam-welded edges in the aforesaid form of construction, is preferably less than 5% of the maximum cross-section dimension of the inner tube.

The invention is illustrated by the accompanying drawings wherein Figure 1 is a diagrammatic section of a double-tube reformer tube assembly and Figure 2 is a section along line II – II of Figure 1.

The reformer tube assembly comprises an outer reformer tube 10 of circular cross-section, closed at its lower end 11, and suspended from a tube-sheet 12. Surrounding tube 10 for the major part of its length is a sheath tube 13, also of circular cross-section, open at both ends and suspended from a second tube-sheet 14 beneath tube sheet 12.

Disposed within tube 10 is an inner tube 15 open at its lower end and having a non-circular cross-section. Tube 15 is made by seam-welding together the long edges of four

elongated strips of metal 16a, 16b, 16c and 16d, each having a substantially uniform thickness but curved across its width so that the outer surface of the welded structure has an inwardly curved, i.e. concave, cross-section. The upper end 17 of tube 15 is flared to a circular cross-section, for ease of connection to a process gas outlet conduit (not shown).

At the lower end of outer tube 10 is disposed a perforate mesh 18 (not shown in Figure 2) which serves as a restraint for the catalyst units.

In use shaped catalyst units, e.g. pellets or rings, are charged to the space 19 above mesh 18 between the inner surface 20 of the outer tube 10 and the exterior, inwardly curved, surface 21 of inner tube 15. Process gas, e.g. a mixture of natural gas and steam, is fed to the space above tube-sheet 12 and passes down through the catalyst-filled space 19, where it undergoes reforming. The reformed process gas passes through mesh 18 and flows up through the space 22 inside inner tube 15 and leaves the reformer tube assembly at the top of tube 15 via the process gas outlet conduit (not shown).

To provide the heat required for the endothermic reforming reaction, a heating gas is supplied to the exterior of the lower end 11 of tube 10 and passes up through the space 23 between the inner surface 24 of the sheath tube 13 and the outer surface 25 of tube 10. This heating gas then leaves sheath tube 13 at the upper end thereof and is collected from the space between tube-sheets 12 and 14.

In use the concave sides of the inner tube are able to reduce their radius of curvature under force from the catalyst units much more easily, i.e. at much lower force, than if the inner tube has a circular cross-section. By equilibrium this results in a lower reaction force on the catalyst units and thus the catalyst units are less liable to breakage. As a result the inner tube becomes progressively inwardly deformed with each expansion/contraction cycle resulting from heating and cooling the assembly. However the resulting decrease in the cross-sectional area of the inner tube has no significant detrimental effect on the gas flow through the inner tube.

Claims.

1. A reformer tube of the double-tube configuration having an outer tube of substantially circular cross-section and an inner tube disposed within the outer tube, said inner tube having a non-circular cross-section.

2. A reformer tube according to claim 1 wherein the inner tube has a maximum cross-sectional dimension that is at least 1.2 times the cross-sectional minimum dimension.

3. A reformer tube according to claim 1 or claim 2 wherein the inner tube has a polygonal cross-section.

4. A reformer tube according to claim 3 wherein the polygon has 4 or 5 sides.

5. A reformer tube according to claim 3 or claim 4 wherein the sides of the polygon are concave.

6. A reformer tube according to any one of claims 3 to 5 wherein the inner tube comprises a plurality of metal strips of substantially uniform thickness seam-welded together along their edges.

7. A reformer tube according to any one of claims 1 to 6 wherein the maximum thickness of the walls of the inner tube is less than 5% of the maximum dimension of the inner tube.

8. A double-tube reformer including reformer tubes according to any one of claims 1 to 7 having shaped catalyst units randomly packed in the space between the inner and outer tubes.

9. The use in a double-tube reformer of an inner tube of non-circular cross-section.

Fig.1.

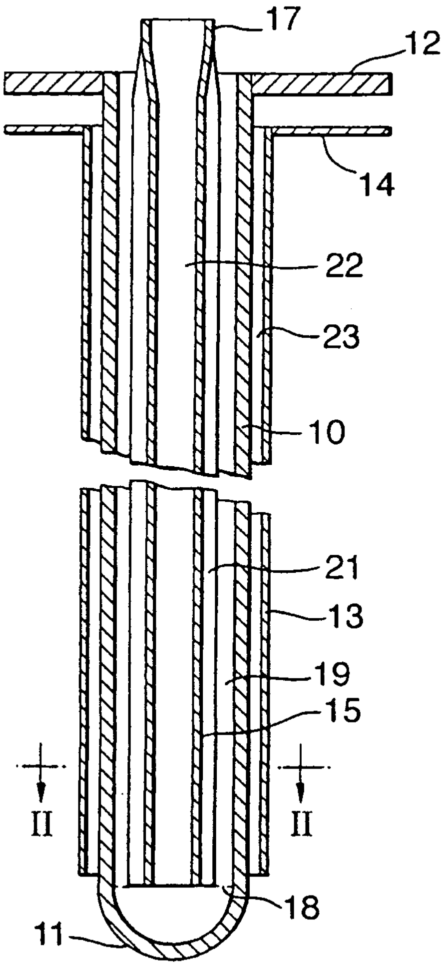
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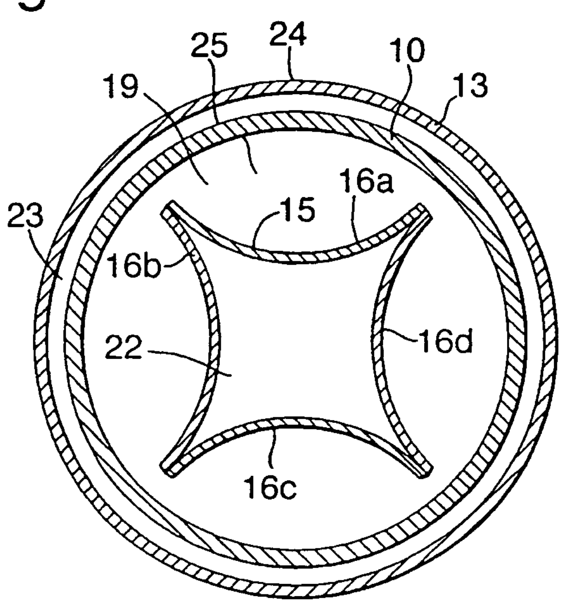
Fig.2.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INTERNATIONAL SEARCH REPORT

International Application No
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A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 B01J8/06 C01B3/38

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 B01J C01B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	FR 841 008 A (FRIED.KRUPP AKTIENGESELLSCHAFT) 9 May 1939 (1939-05-09) page 2, line 12 - line 58 page 3, line 64 - line 73 page 4, line 45 -page 5, line 45 figures 2,3,7,8,12-14	1,4-6
A	DE 24 05 606 A (MARTENS GERHARD) 7 August 1975 (1975-08-07) the whole document	1,9
A	US 2 662 911 A (DORSCHNER OSKAR, WENZEL WILHELM, KAYSER HANS GEORG) 15 December 1953 (1953-12-15) column 14, line 3 - line 37 column 17, line 41 - line 51 figure 17	1,8,9

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
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Name and mailing address of the ISA	Authorized officer
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INTERNATIONAL SEARCH REPORT

Inte 'onal Application No
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Patent document cited in search report		Publication date	Patent family member(s)			Publication date
FR 841008	A	09-05-1939	NONE			
DE 2405606	A	07-08-1975	DE	1901758	A	13-08-1970
			GB	1296193	A	15-11-1972
			SE	366771	B	06-05-1974
			US	3713784	A	30-01-1973
US 2662911	A	15-12-1953	NONE			